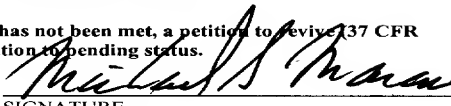


FORM PTO-1390 U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE (REV 11-98)		ATTORNEY DOCKET NUMBER 2119-4203
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		U.S. APPLICATION NO. (If known see 37 CFR 1.51) <b>09/600621</b>
INTERNATIONAL APPLICATION PCT/EP98/08480	INTERNATIONAL FILING DATE 28 December 1998	PRIORITY DATE CLAIMED 20 January 1998
TITLE OF INVENTION LOW PIGMENT CONTENT POLYESTER FILM AND PROCESS FOR MAKING THE SAME		
APPLICANT(S) FOR DO/EO/US Christine ANDREIS; Gusti FEYDER		
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:		
<p>1. <input checked="" type="checkbox"/> This is <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371</p> <p>2. <input type="checkbox"/> This is <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371 (b) and PCT Articles 22 and 39 (1).</p> <p>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</p> <p>6. <input type="checkbox"/> A translation of the International application into English (35 U.S.C. 371(c)(2)).</p> <p>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input type="checkbox"/> have been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p>d. <input checked="" type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)) _____</p> <p>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5))</p> <p><b>Items 11. to 16. below concern document(s) or information included.</b></p> <p>11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98</p> <p>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included</p> <p>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment</p> <p><input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>14. <input type="checkbox"/> A substitute specification.</p> <p>15. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>16. <input type="checkbox"/> Other items or Information:</p>		

U.S. APPLICATION NO. (if known, see 37 C.F.R. 1.51) <b>09/600621</b>		INTERNATIONAL APPLICATION NO. PCT/EP98/08480		ATTORNEY'S DOCKET NO. 2119-4203	
17. <input checked="" type="checkbox"/> The following fees are submitted: <b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5) ):</b> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... \$970.00  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... \$840.00  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2) paid to USPTO ..... \$760.00  International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33 (1) - (4) ..... \$670.00  International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1) - (4) ..... \$96.00  <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>CALCULATIONS</b> <b>PTO USE ONLY</b>	
Surcharge of \$130 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$130.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$	
Total claims	26 - 20 =	6	X \$18.00	\$108.00	
Independent claims	1 - 3 =	0	X \$78.00	\$ 0.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$260.00	\$	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$1,078.00	
Reduction of 1/4 for filing by small entity, if applicable. A Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28).				\$	
<b>SUBTOTAL =</b>				\$1,078.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
<b>TOTAL NATIONAL FEE =</b>				\$1,078.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				\$	
<b>TOTAL FEES ENCLOSED</b>				\$1,078.00	
				<b>Amount to be refunded:</b>	
				<b>charged</b>	
				\$1,078.00	
a. <input type="checkbox"/> A check in the amount of \$ _____ to cover the above fees is enclosed. b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. 13-4500 in the amount of \$ <u>1,078.00</u> to cover the above fees. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-4500. A duplicate copy of this sheet is enclosed.					
<b>NOTE:</b> Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive 37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO:  Morgan & Finnegan, L.L.P. 345 Park Avenue New York, New York 10154 (212) 758-4800 Telephone (212) 751-6849 Facsimile					
SIGNATURE  Michael S. MARCUS NAME					
31.727      July 20, 2000 REGISTRATION NO.      DATE					

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PATENT

Our Docket No. 2119-4203

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Applicant: Christine ANDREIS  
Gusty FEYDER

Serial No.: Unassigned

Group Art Unit: Unassigned

Filed: July 20, 2000

For: LOW PIGMENT CONTENT POLYESTER FILM AND PROCESS FOR  
MAKING THE SAME

Commissioner of Patents  
U.S. Patent & Trademark Office  
Washington, DC 20231

**PRELIMINARY AMENDMENT**

Sir:

Kindly amend the above-identified application prior to examination on the merits as follows:

**IN THE CLAIMS:**

3. (Amended) The film [of] according to claim 1 [or 2] where the ratio is [comprised] between 1 and 10[, preferably 2 and 5].

4. (Amended) The film [of any one of] according to claim[s] 1 [ to 3], wherein the at least one layer[(s)] containing a pigment [contain(s)] contains more than 10 wt% of pigment[, preferably more than 15 wt% of pigment, most preferably more than 20 wt% of pigment].

5. (Amended) The film [of any one of] according to claim[s] 1 [to 4], wherein the at least one layer[(s)] containing a pigment [contain(s)] contains less than 50 wt% of pigment[, preferably less than 40 wt% of pigment].

6. (Amended) The film [of any one of] according to claim[s] 1 [to 5], wherein each layer has a thickness [comprised] in a range of between 1  $\mu\text{m}$  and 200  $\mu\text{m}$ [, preferably between 2 and 50  $\mu\text{m}$ ].

7. (Amended) The film [of any one of] according to claim[s] 1 [to 6], wherein the pigment has a particle size in a range of between 0.01  $\mu\text{m}$  and 5  $\mu\text{m}$ [, preferably between 0.02 and 1.0  $\mu\text{m}$ ].

8. (Amended) The film [of any one of] according to claim[s] 1 [to 7], wherein the layers are coextruded layers.

9. (Amended) The film [of any one of] according to claim[s] 1 [to 8], wherein the pigment is titanium dioxide.

10. (Amended) The film [of any one of] according to claim[s] 1 [to 8], wherein the polyester is PET.

11. (Amended) A process for making the film of [any one of] claim[s] 1 [to 10], comprising the step of coextruding [the various] said layers.

**Please add the following new claims:**

- 13. The film according to claim 3 where the ratio is between 2 and 5.
14. The film according to claim 2 where the ratio is between 1 and 10.
15. The film according to claim 14 wherein the ratio is between 2 and 5.
16. The film according to claim 4, wherein the pigment is present in an amount of greater than 15 wt%.
17. The film according to claim 16, wherein the pigment is present in an amount of greater than 20 wt%.
18. The film according to claim 5, wherein the at least one layer containing a pigment contains less than 40 wt% of pigment.
19. The film according to claim 6, wherein each layer has a thickness in the range of between 2 $\mu$ m and 50 $\mu$ m.

20. The film according to claim 7, wherein the pigment has a particle size in the range of between 0.02 $\mu$ m and 1.0 $\mu$ m.

21. The film according to claim 2, wherein the layers are coextruded layers.

22. The film according to claim 2, wherein the pigment is titanium dioxide.

23. The film according to claim 2, wherein the polyester is PET.

24. A process for making the film of claim 2, comprising the step of coextruding said layers.

25. A process for making the film of claim 3, comprising the step of coextruding said layers.

26. A process for making the film of claim 14, comprising the step of coextruding said layers.

#### REMARKS

Original claims 1-12 according to the amended sheets and newly added Claims 13-25 are pending in the application. Newly added claims 13-25 are fully supported by the original specification and claims. No new matter has been introduced. Examination on the merits is respectfully requested.


**AUTHORIZATIONS:.**

The Commissioner is hereby authorized to charge any additional fees which may be required for the consideration of this preliminary amendment pursuant to 37 CFR §§ 1.16 and 1.17, or credit any overpayment to Deposit Account No. 13-4500, Order No. 2119-4203.

Respectfully submitted,

MORGAN & FINNEGAN, L.L.P.

Dated: July 20, 2000

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LOW PIGMENT CONTENT POLYESTER FILM  
AND PROCESS FOR MAKING THE SAME

## 10 TECHNICAL BACKGROUND OF THE INVENTION

The instant invention relates to a low pigment content polyester film, as well as to a process for making the same.

15 Pigments, especially white pigments, are known additives widely used to impart opacity or specific light transmission properties to films. However, there is currently no film designed for the sole purpose of containing (white) pigments, although there is a need for such films in applications where controlled haze, light  
20 transmission, opacity, etc., are desired final properties.

A film comprised of a mere layer containing pigment is not satisfactory, since the incorporation of fillers, especially pigments, although imparting valuable light transmission properties, presents the drawback of lowering  
25 the mechanical properties of the final film, compared to those of the host polymer prior to incorporation of the filler.

Films are widely known, and afford to combine various properties of different layers that are associated  
30 together. For example, films based on polyester are known.

JP-A-8290539 (to Diafoil Hoechst Co) discloses polyester films where the inner layer comprises fluorescent fine particles and the outer layer comprises white pigment. Said film is said to be adapted as a magnetic recording  
35 medium. The amount of white pigment is however very low.

Similarly, JP-A-8045067 (to Diafoil Hoechst Co) discloses polyester films where the inner layer comprises reclaimed raw material and the outer layer comprises white



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pigment. Said film is said to be adapted as a magnetic card on a pin ball game. The amount of white pigment is however very low.

JP-A-4110147 and JP-A-4110148 (to Diafoil Hoechst Co) disclose polyester films where one of the layers comprises a micropowder in an amount of from 0.1 to 5 wt%. The film has a highly delustered surface and is used for transfer moulding.

JP-A-6000859 (to Diafoil Hoechst Co) discloses polyester films where one of the layers comprises a micropowder in an amount of from 0.001 to 20 wt%, especially from 0.01 to 10 wt%. The film is said to be adapted as a magnetic recording medium, as well as a packing film, a matted film, etc.. All examples make however use of very low amounts of the filler, typically about 1 wt%. Further, the process specified in said patent application is based (i) on extrusion-lamination; (ii) direct feeding with the result of the continuous polymerisation process and (iii) mixing of the filler or additive at the extruder level. This process does not make use of "chips" of polyester, and the feeding of the filler or additive at the extruder level makes it almost impossible to obtain a uniform dispersion of the filler or additive for high amounts, e.g. above 5 wt%.

Although films where one layer contains pigments are known from the above publications, it should be emphasized that these films have not been designed for the sole purposes of containing a white pigment. The problem faced when high amounts of pigments required for imparting high opacity are incorporated is that the mechanical properties of the final film are reduced. It is in fact well established that high opacity and high mechanical properties cannot be found in the same film. Further, it is also well established that high opacity requires high pigment content. Thus there is a need for a film that would show good mechanical properties together with good opacity properties, while minimizing the overall pigment content.

SUMMARY OF THE INVENTION <sup>3</sup>

The invention thus provides such a film, in the form of a film comprising:

(i) at least one polyester layer containing more than 5 wt% of a pigment, and

(ii) at least one polyester layer substantially devoid of pigment,

where the ratio of the respective thickness of the layer(s) devoid of pigment to the pigmented layer(s) is at least 1.

The invention allows to combine the light transmission properties of a layer having a high pigment content together with the traditional mechanical properties of a layer substantially devoid of pigments.

## DESCRIPTION OF EMBODIMENTS OF THE INVENTION.

The polyester used in the invention is any polyester where the major part of it is comprised of any aromatic repeating ester units. The term polyester in this invention refers to a polymer that is obtained by condensation polymerization of an aromatic dicarboxylic acid such as terephthalic acid or 2,6-naphthalene dicarboxylic acid and of an aliphatic terephthalic acid glycol such as ethylene glycol, 1,4-butanediol or 1,4-cyclohexane dimethanol. These polymers, in addition to being homopolymers, may also be copolymers having a third component or several components.

In this case, the dicarboxylic acid component may be, for example, isophthalic acid, phthalic acid, terephthalic acid, 2,6-naphthalene dicarboxylic acid, 4,4'-diphenyldicarboxylic acid, adipic acid, sebacic acid, decanedicarboxylic acid and 1,4-cyclohexane dicarboxylic acid; the oxycarboxylic acid component can be, for example, p-oxybenzoic acid and the glycol component can be, for example, ethylene glycol, diethylene glycol, triethylene glycol, propylene glycol, butanediol, neopentyl glycol, 1,4-cyclohexane dimethanol, polyethylene glycol and polytetramethylene glycol. Examples of such polyesters are polyethylenenaphthalate (PEN), polybutyleneterephthalate (PBT), polyethyleneterephthalate (PET), the latter being the preferred polyester. Mixtures are also possible,

optionally with another polymer different from a polyester. The intrinsic viscosity of the polyester that is used in the invention may vary from e.g. 0.45 to e.g. 0.7, measured in phenoltetrachlorethane at 30°C. The MW may vary within  
5 broad limits, e.g. between 10000 to 30000 g/mol.

Examples of pigments are  $\text{TiO}_2$ ,  $\text{SiO}_2$ ,  $\text{CaCO}_3$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{BaSO}_4$ , carbon black, zeolite, kaolin, the preferred one being  $\text{TiO}_2$ . Mixtures are also possible. Preferably, the pigment is a white pigment. Voids can also be provided  
10 around the particles of pigment, as taught by EP-A-0688814. The pigment used can be the same or different in the pigmented layers, when more than one is used. The pigment has a particle size usually comprised between 0.01 and 5 $\mu\text{m}$ , for example between 0.02 and 1.0 $\mu\text{m}$ .

15 The following description is given with reference to PET and  $\text{TiO}_2$  for the sake of convenience only; it shall not be construed as limited to these embodiments.

The layer containing the pigment will contain at least 5 wt%, preferably at least 10 wt%, especially at least 15  
20 wt% of pigment, most preferably more than 20 wt% of pigment, while the layer substantially devoid of pigment will contain less than 1wt%, and is preferably without pigment.

Upper limits for the content of pigment can be less  
25 than 50 wt% of pigment, preferably less than 40 wt% of pigment.

Thus, ranges for the pigment content are, e.g. 15-50 wt%, preferably 20-40 wt%.

Conventional additives may be incorporated in the  
30 layers of the instant films, in conventional amounts. Examples thereof are anti-oxidant, anti-UV agents, etc.. Mixtures are also possible.

The instant film formed of the various layers can be of various structures and the polyester used can vary from  
35 one layer to another. For example, the inner pigment-free layer and/or the outer pigmented layer(s) can be obtained from starting products containing scrap material, allowing recycling. For example, the inner layer can contain scrap

material, while the pigmented layers do not. The inner layer may even contain scrap material of the final film of the invention, provided the content of pigment is less than 1 wt%. Content of scrap is variable within broad limits known to the skilled man. One pigmented layer can be matte while the other is glossy. Also, the polyester can have a different nature from one layer to another, or they can be the same. All layers can be comprised of crystalline polyester, or all layer can be comprised of amorphous copolyester, or one layer can be crystalline and the other(s) can be amorphous. Especially, the invention provides films where the inner layer is crystalline (i.e. PET) while one or two of the outer layer(s) is amorphous (i.e. copolyester). This allows to combine specific mechanical properties of crystalline polyester and adhesive properties of the copolyester. It can also be foreseen that each layer may be formed of two or more sub-layers; e.g. the outer pigmented layer can be formed of one sub-layer of PET and one sub-layer of copolyester, the first one being in contact with the inner layer.

The instant films can be used as individual layers in further multi-layers films. When necessary, a binder known to the skilled man can be used to incorporate the instant films into a further multi-layers arrangement.

The thickness of each layer can vary within broad limits, for example between 1 and 200 $\mu$ m, preferably between 2 and 50 $\mu$ m. One specific embodiment is the film where the layer substantially devoid of pigment represents the major part of the PET thickness; especially the ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] can be comprised between 1 and 10, preferably 2 and 5.

The invention also provides a process for making the instant films, where the layers are coextruded. Coextrusion operative conditions are within the general knowledge of the skilled man. Other processes such as lamination, calendering, etc. can also be contemplated.

One preferred process makes use of a master-batch, where the pigment represents, e.g. 50wt%. The pellets or

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chips of said master-batch are then mixed with pure polyester pellets or chips and fed into the extruder. The master-batch can also be mixed with chips of recycled (coextruded) films, thus possibly already containing a small amount of pigment. This allows to have a very uniform dispersion of the pigment in the polyester, which would otherwise not be obtainable by direct feeding of the pigment into the extruder.

The following examples are provided for illustrative purposes only and shall not be considered as limiting the scope of the invention.

#### COMPARATIVE EXAMPLES

##### Comparative Example 1.

A resin composition consisting of 95 wt% of PET and 5 wt% of anatase-type titanium oxide was melt extruded at 280°C by a twin-screw extruder through a T shaped die and cooled down on a chill-roll, which was water-cooled at 30°C. Then, the non-drawn sheet was stretched at 80°C by a roll stretching machine at a draw ratio of 3.4 in the MD and then at 130°C by a tenter at a draw ratio of 3.8 in the TD, and fixed at 230°C by heat while relaxing the sheet at a rate of 4%. The resulting film was 20µm thick with the average particle size of the fine titanium oxide particles being less than 0.5µm.

##### Comparative Example 2.

A 20µm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 92.5 wt% of PET and 7.5 wt% of anatase-type titanium oxide is used.

##### Comparative Example 3.

A 20µm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 90 wt% of PET and 10 wt% of anatase-type titanium oxide is used.

##### Comparative Example 4.

A 20µm film was obtained by the same procedure as in comparative example 1 except that a resin composition

consisting of 86 wt% of <sup>7</sup>PET and 14 wt% of anataze-type tinanium oxide is used.

Comparative Example 5.

5 A 40µm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 95 wt% of PET and 5 wt% of anataze-type tinanium oxide is used.

Comparative Example 6.

10 A 40µm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 91 wt% of PET and 9 wt% of anataze-type tinanium oxide is used.

Comparative Example 7.

15 A 12µm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 95 wt% of PET and 5 wt% of anataze-type tinanium oxide is used.

Comparative Example 8.

20 A 12µm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 90 wt% of PET and 10 wt% of anataze-type tinanium oxide is used.

WORKING EXAMPLES.

Example 1.

25 A resin I composition consisting of 95 wt% of PET and 5 wt% of anataze-type titianium oxide and a resin II composition consisting of 100 wt% of PET are melt extruded at 280°C in two twin-screw extruders through a coextrusion-feedblock in such a way that resin I is separated in two melt streams to  
30 form a sandwich structure with resin II as the inner layer. The three layers cast is cooled down on a chill-roll, which was water-cooled at 30°C. Then, the non-drawn 3 layers cast is stretched as in the comparative examples. The resulting film is 15µm thick with a thickness distribution  
35 [PET/TiO<sub>2</sub>]/PET[PET/TiO<sub>2</sub>] of 3.5/8/3.5. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 1.14 and the overall TiO<sub>2</sub> content is the 3 layers film is 2.33 wt%.

Example 2

A 15µm coextruded white film was mobtained by the same procedure as in example 1 except that the composition of resin I was 93% by weight of polyethylene terephthalate and 7% by weight anataze-type TiO<sub>2</sub>. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 1.14 and the overall TiO<sub>2</sub> content in the 3 layer film is 3.27 wt%.

Example 3

A 15µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 91% by weight of polyethylene terephthalate and 9% by weight anataze-type TiO<sub>2</sub>. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 1.14 and the overall TiO<sub>2</sub> content in the 3 layer film is 4.2 wt%.

Example 4

A 15µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 89% by weight of polyethylene terephthalate and 11% by weight anataze-type TiO<sub>2</sub>. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 1.14 and the overall TiO<sub>2</sub> content in the 3 layer film is 5.13 wt%.

Example 5

A 15µm coextruded white film was obtained by the same procedure as in Exazmple 1 except that the composition of resin I was 86% by weight of polyethylene terephthalate and 14% by weight anataze-type TiO<sub>2</sub>. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 1.14 and the overall TiO<sub>2</sub> content in the 3 layer film is 12.25 wt%.

Example 6

A 16µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 81% by weight of polyethylene terephthalate and 19% by weight anataze-type TiO<sub>2</sub>. The thickness ratio [PET-TiO<sub>2</sub>]/PET/[PET-TiO<sub>2</sub>] was 2µm/12µm/2µm. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 3 and the overall TiO<sub>2</sub> content in the 3 layer film is 4.75 wt%.

Example 7

A 15 $\mu$ m coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 74% by weight of polyethylene terephthalate and 26% by weight anatase-type TiO<sub>2</sub>. The thickness ratio [PET-TiO<sub>2</sub>]/PET/[PET-TiO<sub>2</sub>] was 2 $\mu$ m/11 $\mu$ m/2 $\mu$ m. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 2.75 and the overall TiO<sub>2</sub> content in the 3 layer film is 6.93 wt%.

Example 8

10 A 20 $\mu$ m coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 75% by weight of polyethylene terephthalate and 25% by weight anatase-type TiO<sub>2</sub>. The thickness ratio [PET-TiO<sub>2</sub>]/PET/[PET-TiO<sub>2</sub>] was 2 $\mu$ m/16 $\mu$ m/2 $\mu$ m. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 4 and the overall TiO<sub>2</sub> content in the 3 layer film is 5 wt%.

Example 9

20 A 20 $\mu$ m coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 86% by weight of polyethylene terephthalate and 14% by weight anatase-type TiO<sub>2</sub>. The thickness ratio [PET-TiO<sub>2</sub>]/PET/[PET-TiO<sub>2</sub>] was 3.5 $\mu$ m/13 $\mu$ m/3.5 $\mu$ m. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 1.86 and the overall TiO<sub>2</sub> content in the 3 layer film is 4.9wt%.

Example 10

25 A 40 $\mu$ m coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 86% by weight of polyethylene terephthalate and 14% by weight anatase-type TiO<sub>2</sub>. The thickness ratio [PET-TiO<sub>2</sub>]/PET/[PET-TiO<sub>2</sub>] was 3.5 $\mu$ m/33 $\mu$ m/3.5 $\mu$ m. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 4.71 and the overall TiO<sub>2</sub> content in the 3 layer film is 2.45 wt%.

Example 11

35 A 40 $\mu$ m coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 85% by weight of polyethylene terephthalate and 15% by weight anatase-type TiO<sub>2</sub>. The thickness ratio [PET-TiO<sub>2</sub>]/PET/[PET-TiO<sub>2</sub>] was 5 $\mu$ m/30 $\mu$ m/5 $\mu$ m. The overall ratio



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[thickness PET]/[thickness PET+TiO<sub>2</sub>] is 3 and the overall TiO<sub>2</sub> content in the 3 layer film is 3.75 wt%.

#### Example 12

A 20µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 86% by weight of polyethylene terephthalate and 14% by weight anatase-type TiO<sub>2</sub>. The thickness ratio [PET-TiO<sub>2</sub>]/PET/[PET-TiO<sub>2</sub>] was 5µm/10µm/5µm. The overall ratio [thickness PET]/[thickness PET+TiO<sub>2</sub>] is 1 and the overall TiO<sub>2</sub> content in the 3 layer film is 7wt%.

The following properties were measured on some films of comparative examples and some films of working examples.

#### (1) Opacity

Opacity is the hiding power of a non-transparent material. It was measured with a Hunterlab Colorquest spectrophotometer in reflection mode using illuminant III VC (wavelength 400 to 680nm). A black and white reference was taken to determine the opacity given in percent. A completely transparent film is equal to 0% opacity.

#### (2) Whiteness index

Whiteness index is associated with a region or volume in the color space in which objects are recognized as white. The degree of whiteness is measured by the degree of departure from a perfect white (ideal white is bluish). It was measured with a Hunterlab Colorquest spectrophotometer in transmission mode using illuminant III C.

#### (3) Isothermal shrinkage

The size of a 5x5 inch sample was measured before and after heat treatment in an oven at different temperatures, namely 150°C and 200°C. The difference in size is measured and expressed in %, expressing stability.

#### (4) Mechanical properties

Modulus, tensile strength, force at 3% elongation (F3), force at 5% elongation (F5) and elongation at break were determined with an Instron equipment at room temperature.

Tables 1 and 2 give the data for the comparative examples and the working examples, respectively, where Th is thickness, Thcoex is thickness of pigmented layer,

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CoexTiO<sub>2</sub>% is the % of TiO<sub>2</sub> in the pigmented layer, total TiO<sub>2</sub>% is the % of TiO<sub>2</sub> in the film. Table 3 gives mechanical data for films of comparative examples 1 and 2 and working examples 7 and 8.

5

Table 1

Comp. Ex.	Th (μm)	TiO <sub>2</sub> (%)	Opacity %	Whitene ss index	DS 150(%)	DS 200(%)
1	20	5	57.70	-	2.2	8.2
2	20	7.5	64.50	27.3	2.8	9.2
3	20	10	71.7	16.5	2.8	9.2
4	20	14	82.2	-	2.4	8.4
5	40	5	66.7	-	3.2	10.2
6	40	9	81.5	-	3.0	9.4
7	12	5	49.6	-	-	
8	12	10	66.0	-	3.6	10.6

Table 2

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Examp le	Th (μm)	Thcoe x (μm)	Coex TiO <sub>2</sub> %	Total TiO <sub>2</sub> %	Opaci ty %	White ness index	DS 150(%)	DS 200(%)
1	15	3.5	5	2.3	39.30	-	2.0	8.0
2	15	3.5	7	3.3	47.7	-	2.0	7.2
3	15	3.5	9	4.2	51.4	-	1.8	6.8
4	15	3.5	11	5.1	56.8	-	1.8	6.6
5	15	3.5	14	12.3	61.6	-	1.6	6.2
6	16	3.5	19	4.8	72.9	20.0	1.6	6.0
7	15	2	26	6.9	65.0	-	1.8	6.6
8	20	2	25	5	66.10	-	1.8	6.8
9	20	3.5	14	4.9	64.0	29.8	1.4	6.0
10	40	3.5	14	2.5	61.8	-	2.6	8.4
11	40	5	15	3.8	66.9	-	2.2	8.8
12	20	5	14	7	69.0	-	1.2	6.6

12  
Table 3

	Ex. 7	Comp. ex. 2	Ex. 8	Comp. ex. 1
Thickness ( $\mu\text{m}$ )	15	20	20	20
TiO <sub>2</sub> (%)	6,9	7,5	5	5
total	26	7.5	25	5
coex				
Opacity	65	64	66	57
Modulus MD	3669	3865	4370	4069
(Mpa) TD	4992	4580	4832	4325
F3 MD	84	87	90	88
(N/mm <sup>2</sup> ) TD	93	93	94	89
F5 MD	99	101	103	105
(N/mm <sup>2</sup> ) TD	103	104	103	99
Tensile MD	184	201	236	215
(N/mm <sup>2</sup> ) TD	258	224	262	221
Elong. MD	138	113	117	111
(N/mm <sup>2</sup> ) TD	259	224	262	221

By comparing tables 1 and 2, it is possible to  
5 conclude the following:

for a given total TiO<sub>2</sub> content and thickness, opacity  
is higher for the coextruded 3 layer films than for the  
completely filled monolayer films.

for a given opacity, the whiteness index is higher for  
10 the films of the invention than for the completely filled  
monolayer films, evidencing that the coextruded films need  
less pigment.

for a given opacity, the isothermal shrinkage of the  
coextruded films is lower than that of the monolayer film,  
15 evidencing that the films of the invention are more stable.

By analyzing the content of table 3, it is possible to  
conclude that all mechanical values obtained for the  
coextruded 3 layers films are higher than those obtained  
for the monolayer film, evidencing superiority of the  
20 instant films.

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The present invention is not limited by the embodiments described above but shall be construed in accordance with the appended claims.

CLAIMS.

1. A film comprising:

(i) at least one crystalline polyester layer containing  
5 more than 5 wt% of a pigment, and

(ii) at least one crystalline polyester layer substantially  
devoid of pigment,

where the ratio of the respective thickness of the layer(s)  
devoid of pigment to the pigmented layer(s) is at least 1.

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2. The film of claim 1 comprising two outer layers  
containing pigment and one inner layer substantially devoid  
of pigment.

15 3. The film of claim 1 or 2 where the ratio is comprised  
between 1 and 10, preferably 2 and 5.

4. The film of any one of claims 1 to 3, wherein the  
layer(s) containing a pigment contain(s) more than 10 wt%  
20 of pigment, preferably more than 15 wt% of pigment, most  
preferably more than 20 wt% of pigment.

5. The film of any one of claims 1 to 4, wherein the  
layer(s) containing a pigment contain(s) less than 50 wt%  
25 of pigment, preferably less than 40 wt% of pigment.

6. The film of any one of claims 1 to 5, wherein each layer  
has a thickness comprised between 1 and 200 $\mu$ m, preferably  
between 2 and 50 $\mu$ m.

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7. The film of any one of claims 1 to 6, wherein the  
pigment has a particle size comprised between 0.01 and 5 $\mu$ m,  
preferably between 0.02 and 1.0 $\mu$ m.

35 8. The film of any one of claims 1 to 7, wherein the layers  
are coextruded layers.

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9. The film of any one of claims 1 to 8, wherein the pigment is titanium dioxide.

10. The film of any one of claims 1 to 8, wherein the polyester is PET.

11. A process for making the film of any one of claims 1 to 10, comprising the step of coextruding the various layers.

10 12. The process of claim 11, comprising the step of using a master-batch.

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## DECLARATION and POWER OF ATTORNEY

As a below-named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**Low Pigment Content Polyester Film and Process of Making the Same**

the specification of which is attached hereto unless the following box is checked:

☒ was filed on **20 July 2000** as U.S. Application No. **09/600,621** or PCT International Application No. \_\_\_\_\_

and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is known to me to be material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Application No.	Country	Filing Date	Priority Claimed (Yes/No)
PCT/EP98/08480	PCT	12/28/98	Yes
98400104.0	EP	01/20/98	Yes

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States Provisional Application(s) listed below,

U.S. Provisional Application No. \_\_\_\_\_

U.S. Filing Date \_\_\_\_\_

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International Application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application or PCT International Application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is known to me to be material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application No.	Filing Date	Status (patented, pending or abandoned)

**POWER OF ATTORNEY:** I hereby appoint the following attorney(s) and/or agent(s) the power to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:Name: **ANDREW G. GOLIAN**Registration No.: **25,293**

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☐ Additional Inventors are being named on separately numbered sheets attached hereto.

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	<b>Filing Date</b>	20 July 2000
	<b>First Named Inventor</b>	Andreis
	<b>Group Art Unit</b>	Unknown
	<b>Examiner Name</b>	Unknown
	<b>Attorney Docket Number</b>	2119-4203

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